PB82-132556

Size and Composition of Visibility-Reducing Aerosols in Southwestern Plumes

Washington Univ.
St. Louis, MO

Prepared for

Environmental Sciences Research Lab.
Research Triangle Park, NC

1980

U.S. Department of Commerce
National Technical Information Service
NTIS

TECHNICAL REPORT DATA (Please read Instructions on the reverse before completing)		
1. REPORT NO. EPA-600/J-80-357	2. Journal Article	3. RECI P882-132556
4. TITLE AND SUBTITLE: SIZE AND COMPOSITION OF VISIBILITY-REDUCING AEROSOLS IN SOUTHWESTERN PLUMES		5. REPORT DATE: October 1981 6. PERFORMING ORGANIZATION CODE: 2337054
7. AUTHOR(S): Edward S. Macias, Donald L. Blumenthal, Jerry A. Anderson and Bruce K. Cantrell		8. PERFORMING ORGANIZATION REPORT NO.
9. PERFORMING ORGANIZATION NAME AND ADDRESS: Department of Chemistry Washington University St. Louis, MO 63130		10. PROGRAM ELEMENT NO.: CABN1D/04-0543 (FY-81) 11. CONTRACT/GRANT NO.
12. SPONSORING AGENCY NAME AND ADDRESS: Environmental Sciences Research Laboratory - RTP, NC Office of Research and Development U.S. Environmental Protection Agency Research Triangle Park, N.C. 27711		13. TYPE OF REPORT AND PERIOD COVERED 14. SPONSORING AGENCY CODE: EPA/600/09
15. SUPPLEMENTARY NOTES: REFERENCE: Annals of the New York Academy of Sciences, 338:233-257, May 15, 1980.		

16. ABSTRACT

The southwestern United States desert and mountain areas are generally characterized by very good visibility. Until recent years, scenic vistas of natural landmarks and mountains with a visual range of over 100 miles were common. These vistas have been considered a major resource of the Southwest, and numerous national parks, forests, and monuments have been created to preserve them. The tourism resulting from the Southwest's scenery has played a major role in the economy of the region.

In this paper, we describe the results of the fall 1977 aircraft measurements of Project VISTTA. The Southwest visibility-reducing aerosol is characterized with regard to spatial distribution, particle size distribution, and chemical composition as a function of size. The emissions from a copper smelter and a coal-fired power plant in the region, as measured in elevated plumes, are characterized with regard to chemical composition as a function of size. The contribution of various species to light scattering is also quantitatively assessed.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group

REPRODUCED BY
NATIONAL TECHNICAL
INFORMATION SERVICE
U.S. DEPARTMENT OF COMMERCE
SPRINGFIELD, VA 22161

18. DISTRIBUTION STATEMENT	19. SECURITY CLASS (This Report)	21. NO. OF PAGES
RELEASE TO PUBLIC	UNCLASSIFIED	
	20. SECURITY CLASS (This page) UNCLASSIFIED	22. PRICE

EPA-600/J-80-357
Journal Article

SIZE AND COMPOSITION OF VISIBILITY-REDUCING AEROSOLS IN SOUTHWESTERN PLUMES*

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INTRODUCTION

The southwestern United States desert and mountain areas are generally characterized by very good visibility (>100 km). Until recent years, scenic vistas of natural landmarks and mountains with a visual range of over 100 miles were common. These vistas have been considered a major resource of the Southwest, and numerous national parks, forests, and monuments have been created to preserve them. The tourism resulting from the Southwest's scenery has played a major role in the economy of the region.

As the population of the region has grown, there has been an apparent reduction in the average visibility.[1] Residents of the Southwest are concerned about this loss and point to various anthropogenic activities as the cause. This concern has led to the August 1977 amendments to the Clean Air Act, which contain a section stating "as a national goal the prevention of any future, and the remedying of any existing, impairment of visibility in mandatory Class I Federal areas which impairment results from man-made air pollution."§ Previously, however, comprehensive studies had not been performed to quantify and determine the cause of the current problem or to provide the basis for a control strategy to keep the problem from getting worse.

Visibility impairment in the atmosphere is due primarily to the presence of small particles, which scatter and absorb light, and, secondarily, to NO_2, which absorbs blue light.[2] In the Southwest, a regional decrease in visibility is probably due to the long range transport of aerosol or gaseous precursors into unpopulated pristine areas. A second possible cause of regional visibility impairment may be windblown dust. Both types of particles may be of anthropogenic origin. Some of the varied activities that

*Supported by the Federal Interagency Energy/Environment Research and Development program, through a contract with the EPA-ESRL Office of Regional Studies.
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‡Work performed while on leave at California Institute of Technology, Pasadena, Calif.
§Section 169A of the Clean Air Act.

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may contribute to the decline in visibility are: increased population and the resulting increase in automotive emissions; emissions from power plants; emissions from smelters; disturbance of the topsoil and desert pavement due to agriculture, grazing, and off-road vehicles; and mining activities.

Project VISTTA,¶ sponsored by the EPA Environmental Science Research Laboratory in Research Triangle Park, North Carolina, has been initiated to determine the sources of visibility reduction in the Southwest and to help resolve the potential conflict between the congressional mandate for improved visibility and the need for increased energy production.

In this paper, we describe the results of the fall 1977 aircraft measurements of Project VISTTA. The Southwest visibility-reducing aerosol is characterized with regard to spatial distribution, particle size distribution, and chemical composition as a function of size. The emissions from a copper smelter and a coal-fired power plant in the region, as measured in elevated plumes, are characterized with regard to chemical composition as a function of size. The contribution of various species to light scattering is also quantitatively assessed.

Experimental Description

The initial exploratory field program of Project VISTTA was carried out October 1–10, 1977. Measurements were taken within the mixing layer (<500 m above ground level), using the instrumented MRI Beechcraft Queen Air aircraft. This twin-engine aircraft has minimum and maximum sampling speeds of 180 and 290 km/hr, respectively. With a full instrument complement, the maximum sampling time is over four hours. The aircraft was equipped to continuously measure and record on magnetic tape light scattering coefficient, Aitken nuclei count, ozone, sulfur dioxide, nitrogen oxides, temperature, dew point, turbulence, pressure (altitude), and navigational parameters. Aerosol size distribution measurements were made over a range of 0.01 to 30 μm, using three size-selective aerosol sensors. An electrical aerosol analyzer and optical particle counter were used in conjunction with an automated "grab bag" sampling system designed to collect and hold a discrete static air sample for analysis. An axial scattering probe was operated in the free stream near the nose of the aircraft for particle size measurements in the range of 2 to 30 μm. Multistage impactor and two-stage filter sampler samples were also collected, in order to determine the elemental composition as a function of size. The details of the aircraft instrumentation are given in Table 1. Visual range measurements were carried out by viewing distant landmarks and verified by optical photography.

The multistage impactor, a Lundgren-type impactor specially modified for aircraft sampling by investigators at the University of California at Davis (UCD), was mounted in the nose assembly of the aircraft. The impactor stages had 50% collection efficiency for particle diameters of 4, 2, 1, and 0.5 μm. Impactor samples were collected on greased Mylar film. A Nuclepore final filter was used to collect all particles not collected on the four impaction stages. The impactor samples were analyzed by particle-induced x-ray emission (PIXE)[4] at the Crocker Nuclear Lab

¶VISTTA is an acronym for Visibility Impairment due to Sulfur Transport and Transformation in the Atmosphere.

cyclotron at UCD. This analysis gives elemental concentration of elements with atomic numbers of 13 (Al) or greater.

The fine particle samples from the TWOMASS two-stage sampler[5] were analyzed by flash vaporization–flame photometric detection (FV–FPD)[6] by J. D. Husar and Associates (St. Louis, Missouri).

The field program consisted of seven sampling days. The experimental plan included flights of the region on three full days and parts of one other, flights in the

TABLE I
BEECHCRAFT QUEEN AIR INSTRUMENTATION

Measured Parameter	Instrument
Aerosols—Integral Size:	
Light Scattering	MRI 1562 Nephelometer (modified by UW for high sensitivity)
Aitken Nuclei	Environment One Condensation Nuclei Monitor
Aerosols—Differential Size:	
Particle Diameter;	
0.01–1.0 μm	TSI 3030 Electrical Aerosol Size Analyzer (EAA)
0.5–4.0 μm	Royco 218 Optical Particle Counter (OPC)
2.0–30.0 μm	Particle Measurement Systems Axial Scattering Probe (ASP)
Aerosol Samples:	
Two Stage Sampler	MRI TWOMASS Sampler
Multistage Impactor	UCD Impactor System
Gases:	
Ozone	REM 612 O_3
Sulfur Dioxide	Meloy 285 Total Sulfur Monitor
Nitrogen Oxides	Monitor Labs 8440 NO–NO_x
Other:	
Dew Point	Cambridge Systems 137
Temperature, Turbulence, Altitude, Indicated Air Speed	MRI Airborne Instrument Package
Position: VOR, DME	Aircraft Navigation System
Visual Range	Optical Photography

plume of a copper smelter (San Manuel smelter) on three days, and flights in the plume of a coal-fired power plant (Mohave power plant) on one day.

The regional flights were designed to characterize the visibility-reducing aerosol on flight paths of over 300 km. On two days, the total flight paths were over 1000 km. All flights were within the mixing layer. However, occasional spirals were made to define the mixing layer structure. The visibility along these paths was documented with a high-sensitivity nephelometer, a camera, and with visual range measurements from the aircraft to prominent landmarks.

Smelter plume flights were carried out at several downwind distances up to 127 km from the San Manuel smelter. Flights in the 1580 MW Mohave power plant plume were performed at distances as far as 62 km downwind of the plant.

The plume flight patterns were designed to provide a detailed characterization of the plume at discrete distances downwind from the source. At each distance, horizontal traverses were made through the plume normal to the plume axis at several elevations. Traverses were long enough to include background air on either side of the plume. Vertical spirals were also performed to help characterize the plume structure. In addition to cross-sectional plume traverses, orbital flights within the plume of up to one hour were carried out at each distance, to gather heavily loaded impactor and filter samples. Orbits upwind of the source were flown to gather background samples.

Single theodolite pilot balloon (pibal) releases were carried out at various times on every sampling day, to determine wind direction and speed as a function of altitude in the sampling area.

Results and Discussion

Southwest Background Measurements

Spatial Distribution

The character of visibility-reducing aerosols in the Southwest was determined in long regional flights, in order to measure a regional background. These flights were made between or upwind of large point sources of pollutants, such as urban areas and power plants. All flights were at a constant altitude to within ±25 m. On October 5, 1977, flights were made between Flagstaff (Arizona)–Tuba City (Arizona)–Island in the Sky (Utah)–Farmington (New Mexico)–Grants (N. Mex.)–Flagstaff, as shown in Figure 1. The flights on October 9 followed the same 1080-km path in the reverse direction. Shorter flights in the region (~260 km) were flown on October 8 and 10. On all days, the visibility and air quality were quite good (average visual range – 140 km). This resulted in very low values for all measured aerosol and gas parameters except ozone, which was present in concentrations representative of clean air [36 ppb (O_3)]. Therefore, only data from the longer flights on October 5 and 9, which have more meaningful averages, are discussed in this paper. The averages for each day were determined by averaging over each segment of the flight and then averaging the flight segment averages. The NO, NO_x, and SO_2 concentrations were below the sensitivity of the monitors throughout the region (<5 ppb NO and NO_x, <1 ppb SO_2). The b_{sp} values, although quite low, were detectable with the aircraft instrumentation.

The spatial distribution of the visibility-reducing aerosol is most easily determined from the frequency distribution of the light scattering coefficient due to particles, b_{sp}, as shown in Figure 2. These values are determined from the 10-sec average nephelometer readings, with the Rayleigh scattering contribution to the scattering coefficient at the flight altitude subtracted.

The average value of b_{sp} on both sampling days was rather low, ranging from $(14 \pm 1) \times 10^{-6}$ m^{-1} on October 9 to $(17 \pm 3) \times 10^{-6}$ m^{-1} on October 5. Although

these values are low, t
[b_{sp}(min) ≃ 5 × 10^{-6} m^{-}
sampling altitude (~ 11
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made. The average meas
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VISTTA data can be co

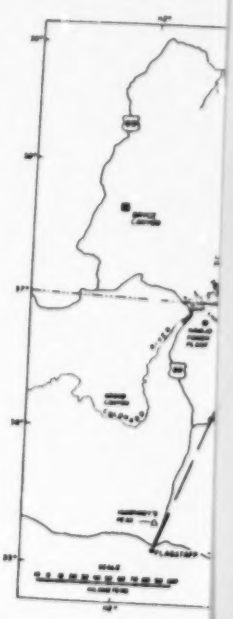

Figure 1. Flight path of V
path was ~1080 km.

The coefficient of varia
and 18% on October 5 ov
aerosol was homogeneous

Size Distribution

The average size distri
aircraft during the Octobe

these values are low, they are above the minimum sensitivity of the instrument [$b_{sp}(\min) \simeq 5 \times 10^{-6}$ m^{-1}]. The Rayleigh scattering, or clean air, contribution at the sampling altitude ($\sim 11 \times 10^{-6}$ m^{-1}) was, on the average, 42% of the total light scattering coefficient during the very clear days on which these regional flights were made. The average measured visual range in Mesa Verde and other national parks in the region in 1976[7] was about equal to the average visual range calculated from the scattering coefficient on the VISTTA sampling days (~150 km). Therefore, the VISTTA data can be considered typical for the region.

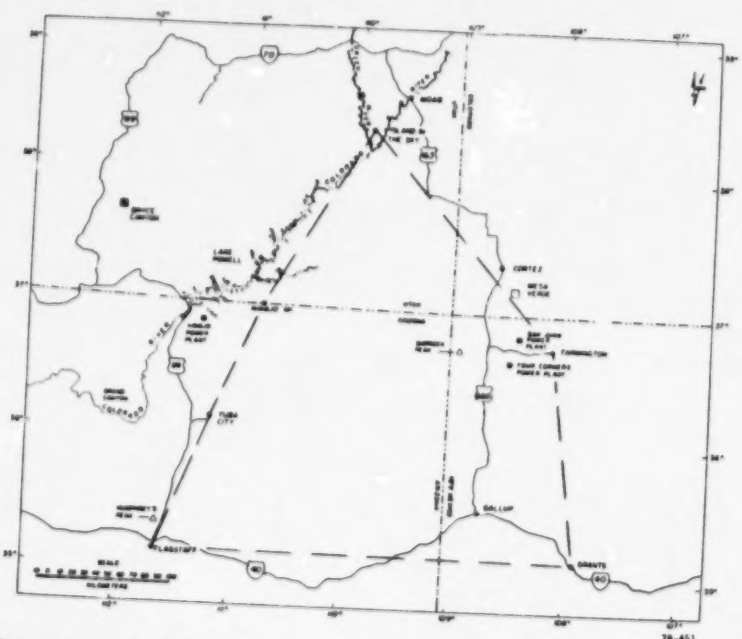

FIGURE 1. Flight path of VISTTA regional flights on October 5 and 9, 1977. The entire flight path was ~1080 km.

The coefficient of variation of the b_{sp} frequency distribution was 7% on October 9 and 18% on October 5 over the entire 1080-km flight path, which indicates that the aerosol was homogeneous on both days.

Size Distribution

The average size distribution of background Southwest aerosol measured on the aircraft during the October 9 regional flight is shown in FIGURE 3. The distribution is

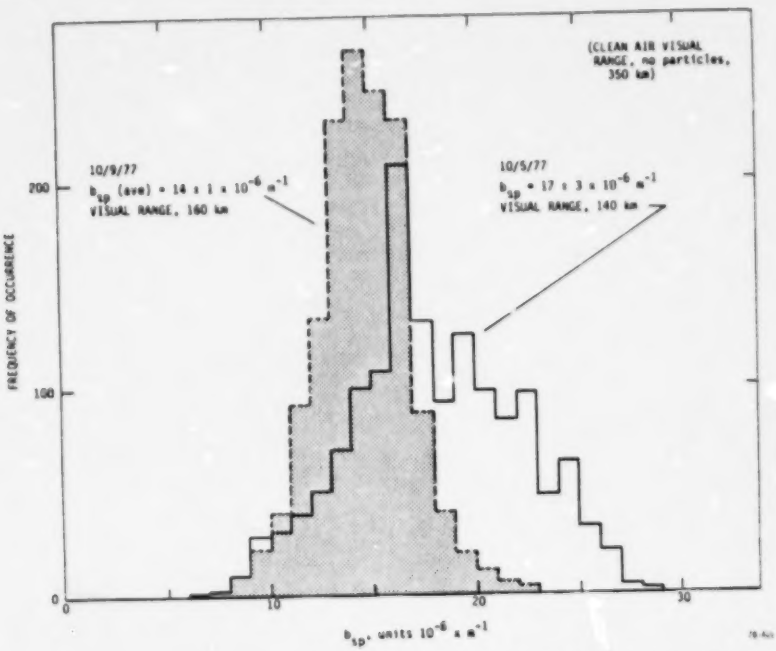

FIGURE 2. Frequency distribution of the light scattering coefficient due to particles, b_{sp}, measured in regional flights on October 5 and 9, 1977. Measurements were averaged over 10 sec (1 km) for the 1080-km flight path for each day. The b_{sp} averages on October 5 and 9 were 17 and 14 × 10^{-6} m^{-1}, with standard deviations of 3 and 1 × 10^{-6} m^{-1}, respectively, indicating that the aerosol was homogeneous on these days.

bimodal. The aerosol size distribution data were reduced, using procedures described elsewhere.[8] The size distribution data were collected every five minutes during the entire flight. All measured size distributions were averaged before extracting integral parameters using a log-normal approximation technique. There is some question about altitude effects on these instruments, which may lead to errors in the size distribution measurements. However, we do not think these errors are large enough to alter the conclusions of this paper. We are presently looking into the magnitude of this flow rate effect. The fine particle, or accumulation, mode had a geometric volume mean size, D_{pv} of 0.25 μm and a geometric standard deviation, σ, of 2.0. The coarse mode geometric volume mean size is 5.5 μm, with a σ of 2.2. The volume concentration of particles is 2.6 $\mu m^3/cm^3$ and 2.5 $\mu m^3/cm^3$ for the fine and coarse modes, respectively. The accumulation mode mean size is slightly smaller than the continental average of 0.3 μm.[9] However, the coarse particle mean size is the same, within experimental error, as the continental average of 6 μm expected for particles from similar sources, such as windblown dust.

Elemental Composition

The elemental composition of the visibility-reducing aerosol in the Southwest was determined from impactor and filter samples collected on regional flights October 5 and 9, 1977. As discussed previously, the aerosol was quite homogeneous throughout the region during this sampling, making a regional average meaningful. The elemental concentrations, divided into fine particles ($D_p < 1\mu m$) and coarse particles ($D_p > 1$ μm), averaged for all samples collected on October 5 and 9 are given in TABLE 2. The rationale for the separation at 1 μm is based on the high scattering efficiency for particles with diameters less than 1 μm. Furthermore, particles in each of these size ranges generally have different chemical compositions. All other elements not listed were below the minimum detectable limits. The elemental concentration of fine particle sulfur was measured by FV–FPD analysis of TWOMASS samples. All other data are from PIXE analysis of impactor samples. There was some discrepancy between the values from the two techniques, but the FV–FPD values appeared more reliable. The coarse particle fraction is dominated by crustal elements, such as silicon, aluminum, calcium, and iron. Sulfur, silicon, and potassium are predominant elemental species in the fine particles.

The size distribution of individual elemental species, determined from regional impactor samples collected on the October 5 and 9 regional flights, is shown in FIGURE 4. Aluminum, calcium, and iron all show very similar size distributions, with the highest concentration of each element in particles with diameters greater than 4 μm. The concentration on the final filter ($D_p < 0.5$ μm) was below the minimum detectable limit for all these elements. Silicon has a size distribution similar to aluminum down to

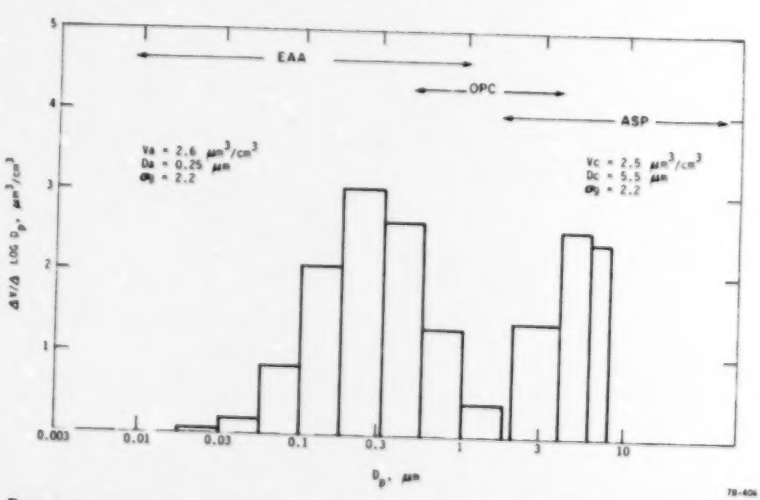

FIGURE 3. Average size distribution of background Southwest aerosol, measured on the October 9, 1977 regional flight.

TABLE 2

AVERAGE ELEMENTAL CONCENTRATION OF AEROSOL IN THE SOUTHWEST REGION, OCTOBER 5 AND 9, 1977

Element*	Fine Particles† ($\mu g/m^3$)	Coarse Particles‡ ($\mu g/m^3$)
Al	0.01	0.28
Si	0.72	1.12
S	0.68	0.06
Cl	0.11	0.01
K	0.21	0.09
Ca	0.03	0.24
Ti	0.08	0.01
V	0.04	0.01
Cr	—	0.01
Fe	0.004	0.16
Zn	0.03	0.003
Zr	—	0.02
Pd	0.02	0.01
Ba	0.10	0.01
Pb	0.09	0.004

*The following elements were detected in only a few samples, and, therefore, it is not possible to determine a meaningful average concentration: Fine Particles—Cu, Mn, Ni, Rb, Co, Hg, Mo, Pt. Coarse Particles—Cu, Mn, Se, Ni, Rb, Co, Mo, Br, Pt.

†Fine particle ($D_p < 1$ μm) concentrations were determined from the sum of impactor stage 4 and the final filter. Sulfur values were determined from TWOMASS samples.

‡Coarse particle ($D_p > 1$ μm) concentrations were determined from the sum of impactor stages 1, 2, and 3.

1 μm; below that diameter, the concentration of silicon increases. The concentration of small particle silicon ($D_p < 0.5$ μm) is nearly equal to the large particle value ($D_p > 4$ μm). These data indicate that silicon is a major contributor to the fine particle mass and, as discussed below, to visibility reduction. However, subsequent measurements have shown lower silicon concentrations. From the source characterization determined in this work, fine particle silicon results from the condensation of volatile silicon emitted from high-temperature combustion sources.

Chemical Composition

The total aerosol mass was not measured, but it can be estimated from the measured elemental concentrations, and the oxygen concentration estimated with assumptions of the chemical form of the measured elements. The sulfur can be expressed as sulfate because the FV–FPD method is specific for water-soluble sulfate.[8] It is also assumed that ammonium is the only ion associated with the sulfate. Other elements are assumed to exist as oxides. This leads to the mass balance listed in TABLE 3 and FIGURE 5. The mass estimates may be low due to unmeasured elements, particularly those with $Z < 13$. The total mass in each size range can also be estimated from the aerosol volume determined by integrating each mode in the differential size distribution and by estimating the aerosol density from the chemical composition. A

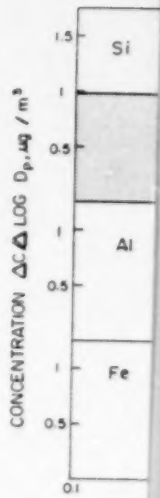

FIGURE 4. Averag
impactor samples coll
to Al, Ca, and Fe do

CHEMICAL SPECIES

Compound
Al_2O_3
SiO_2
$(NH_4)_2SO_4$
K
CaO
TiO_2
Fe_2O_3
ZnO
PbO
Other (in normal oxide form)
Total

*The percent cont
concentrations.

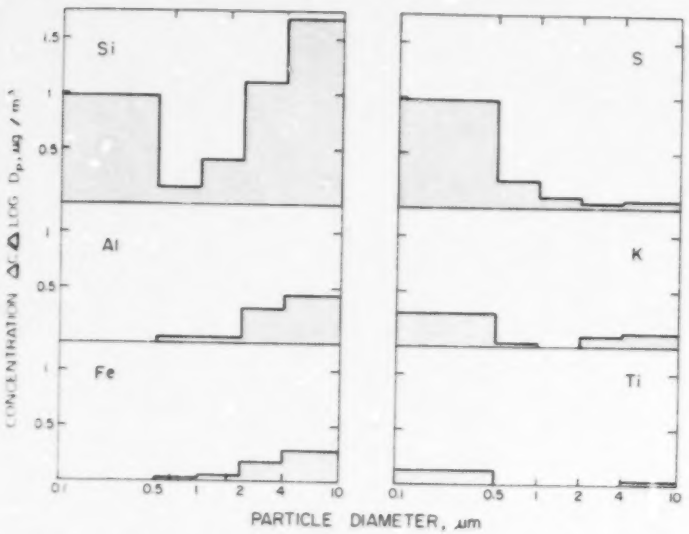

FIGURE 4. Average size distribution of individual elemental species, determined from impactor samples collected on October 5 and 9 regional flights. Si has a size distribution similar to Al, Ca, and Fe down to 1 μm; below that diameter, the concentration of Si increases.

TABLE 3

CHEMICAL SPECIES BALANCE FOR THE SOUTHWEST REGION—OCTOBER 5 AND 9, 1977

Compound	Fine Particles		Coarse Particles	
	Concentration ($\mu g/m^3$)	Contribution* (%)	Concentration ($\mu g/m^3$)	Contribution (%)
Al_2O_3	0.02	0.4	0.53	13.4
SiO_2	1.54	29.2	2.40	60.6
$(NH_4)_2SO_4$	2.81	53.3	0.25	6.3
K	0.21	4.0	0.09	2.3
CaO	0.04	0.8	0.34	8.6
TiO_2	0.13	2.5	0.02	0.5
Fe_2O_3	0.006	0.1	0.23	5.8
ZnO	0.04	0.8	0.004	0.1
PbO	0.10	1.9	0.004	0.1
Other (in normal oxide form)	0.37	7.0	0.09	2.3
Total	5.27	100.0	3.96	100.0

*The percent contribution is based on the mass estimated from the sum of the chemical concentrations.

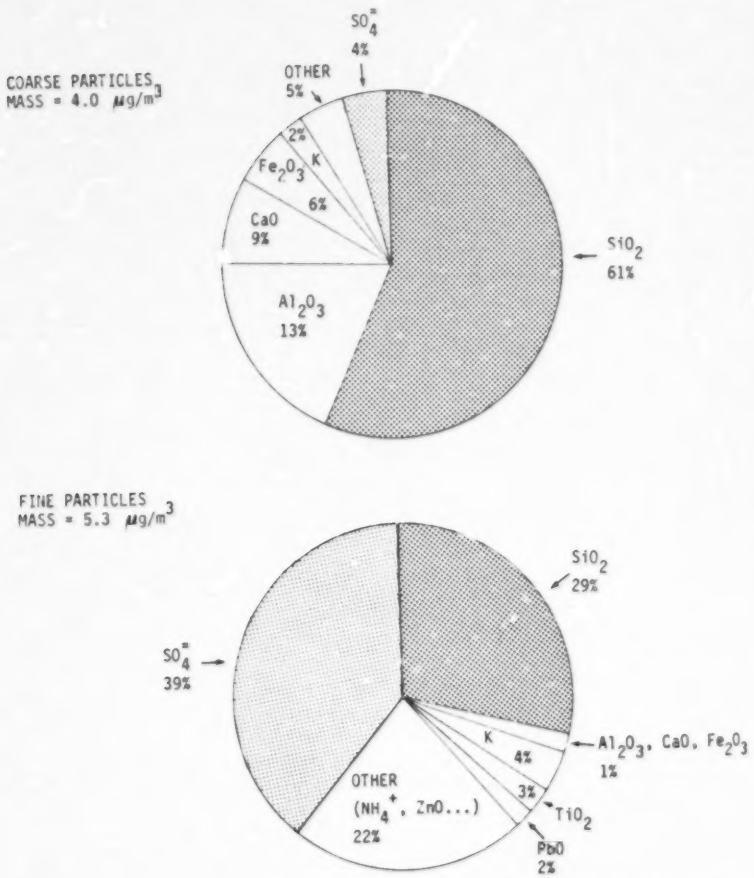

FIGURE 5. Size-fractionated mass balance of Southwest background aerosol, measured on regional flights on October 5 and 9, 1977. The concentration of individual compounds was determined from the measured elemental concentration and assumed chemical form. The total aerosol mass in each size fraction was determined from the sum of the individual chemical components. Comparison with aerosol volume measurements indicates that the mass estimated from composition measurements represents 93% and 64% of the fine and coarse particle mass, respectively.

comparison of these two approaches indicates that the elemental mass balance accounts for ~93% and ~64% of the fine and coarse particle mass derived from volume, respectively. The assumptions of chemical form, although somewhat arbitrary, lead to a conservative estimate of the total mass. For example, iron has been assumed to be present exclusively as Fe_2O_3, when in reality it is likely that FeO is

present as well. This is the case for crustal material.[10] Comparison of fine particle mass and b_{sp} allows a further check on the mass estimate. Extensive measurements in the western United States indicate that the fine particle mass/b_{sp} ratio is normally 0.32 to 0.34 g/m.[11] The ratio for the VISTTA regional data is 0.34 g/m, which is a further indication that the fine particle mass estimate is reasonable.

The total measured aerosol mass in both size ranges (9.23 $\mu g/m^3$), determined from the sum of the concentration of each compound, is lower than the average total suspended particulate value (~20 $\mu g/m^3$) measured in several national parks in the region.[1] It appears from volume and measurements that this discrepancy is not primarily due to fine particle mass estimates. It is more likely that the coarse particle mass estimate is low due to unmeasured constituents. Furthermore, coarse particles may be diminished due to settling at 300 to 500 m above the ground, where the samples were collected.

Probably the most important species unaccounted for are nitrates, carbon compounds, and water. The average annual concentration of nitrate in the nonurban Southwest region was about 20% of the sulfate value in the early 1970s, as measured in Grand Canyon and other remote southwestern national parks by the National Air Surveillance Network.[7] These values may be subject to error due to artifact sulfate and nitrate effects on filters. Very few data exist on the concentration of carbon in Southwest ambient aerosols, but it is a significant component of eastern urban aerosols.[12]

Enrichment Factors

The predominant species in the coarse particles are elements found in high concentration in soil and crustal material. Elemental enrichment factors, given in TABLE 4, help to characterize the source of these particles. These factors are calculated as follows:

$$\text{Enrichment Factor (EF)} = \frac{(X/\text{Al})\ \text{Aerosol}}{(X/\text{Al})\ \text{Crust}}$$

Aluminum was chosen as the reference element because its major source is likely to be soil. However, aluminum is also present in fly ash from coal combustion.[13] Enrichment factors >1 indicate an enrichment of that element relative to crustal abundances; values <1 indicate a depletion relative to crust. Because of the uncertainty in the data, EF values between 0.5 and 2 indicate no significant difference from crustal abundance. It can be seen from TABLE 4 that Si, K, Ca, Ti, V, and Fe are all near the crustal abundance in the coarse particles relative to Al. These elements represent 93% of measured coarse particle mass. The other coarse particle species are enriched, indicating an anthropogenic source. This analysis is complicated by the fact that many of these elements are present in coal fly ash, with abundances similar to crustal material.

All fine particle species except iron are enriched relative to the crust (normalized to aluminum), with enrichment factors ranging from 7 to 3×10^7. The enriched fine particle species represent 99.5% of the measured fine particle mass, which indicates that the vast majority of fine particles are not of crustal origin.

TABLE 4
ENRICHMENT FACTORS

Element	Enrichment Factor: Fine Particles ($D_p \leq 1$ μm)		Enrichment Factor: Coarse Particles ($D_p \geq 1$ μm)	
Southwest Background				
Al	1.0	Crustal Abundance	1.0	Crustal Abundance
Fe	0.7		0.9	
Si	21		1.2	
K	66		1.9	
Ca	7		0.7	
Ti	1×10^2		67	
S	2×10^4		23	
Cl	7×10^3	Enriched Relative to the Crust	29	Enriched Relative to the Crust
V	2×10^3		13	
Cr	—		35	
Zn	4×10^3		3×10^3	
Zr	—		7	
Pd	2×10^7		3×10^3	
Ba	2×10^3		88	
Pt	3×10^7			
Pb	6×10^4			
San Manuel Smelter 60 km Downwind (10/4/77)				
Al	1.0		1.0	
Si	32		0.9	
K	1×10^2		1.0	Crustal Abundance
Ca	—		2	
Ti	37		0.3	
Mn	—	Enriched	0.9	
Fe	3		0.9	
S	7×10^4		—	
Cr	2×10^4		26	Enriched
Cu	—		46	
Se	4×10^7		—	
Mohave Power Plant 32 km Downwind (10/8/77)				
Al	1.0	Crustal Abundance	1.0	
Si	2		0.7	
K	—		0.5	Crustal Abundance
Ca	—		2	
Ti	—		0.5	
Fe	—		0.6	
S	2×10^3	Enriched	—	
Cr	—		28	Enriched
Ni	4×10^3		7	
Pb	—		128	

Light Scattering Budget

The detailed contributions to light scattering as a function of particle size can be determined from Mie calculations[14] of the light scattering efficiency and the measured particle size distribution. The sum of the calculated b over all particle sizes can be compared to the measured light scattering coefficient from the integrating nephelometer. Mie scattering calculations, combined with particle size and composition

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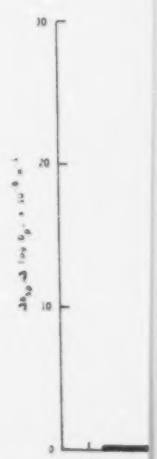

FIGURE 6. Lig
October 9, 1977.
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SiO_2 (29%).

measurements, permit the determination of the individual contribution to visibility reduction for particles of a given composition and size (light scattering budget). This approach can also serve as a test of the consistency of the various measurements.[15] The following expression was evaluated in the scattering coefficient calculation:

$$\bar{b}_{sp} = \int G(D_p) \frac{dV}{d \log D_p} d \log D_p$$

where G is the light scattering per unit volume of aerosol. The quantity G is calculated using Mie scattering functions.

The contributions to the light scattering coefficient as a function of particle size for the average Southwest regional aerosol (October 9, 1977), calculated from the average regional aerosol size distribution, are given in FIGURE 6. A refractive index, $m = 1.54$, was used in this calculation, which is slightly lower than the mass-weighted values of 1.57 and 1.65 for fine and coarse particles, respectively, calculated from aerosol composition measurements. The calculation was done for light of 525 nm wavelength. The sum over all calculated contributions yields:

$$\sum b_{sp} = (16 \pm 2) \times 10^{-6}\ m^{-1}$$

which agrees well with the average value of light scattering due to particles, measured with the integrating nephelometer (with the contribution of Rayleigh scattering due to gases removed), of:

$$b_{sp}(\text{measured}) = 14 \pm 1 \times 10^{-6}\ m^{-1}$$

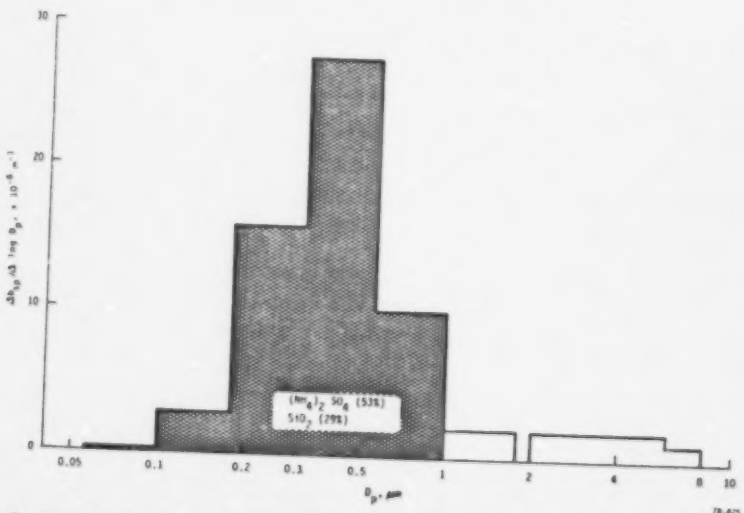

FIGURE 6. Light scattering contribution as a function of size for the Southwest region, October 9, 1977. The fine particle contribution, which represents 93% of the total b_{sp}, is shaded in the figure. The major components of the fine particles ($D_p \leq 1\ \mu m$) are $(NH_4)_2SO_4$ (53%) and SiO_2 (29%).

TABLE 5

LIGHT SCATTERING BUDGET

Component	D_p (μm)	b_{scat} ($\times 10^{-6}$ m^{-1})	Contribution to Total b_{scat} (%)
Southwest Region 10/9/77 (Visual Range ~160 km)			
Air Molecules	—	11	44
$(NH_4)_2SO_4$*	0.1 to 2.0	7	28
SiO_2†	0.1 to 1.0	4	16
Other Compounds‡	0.1 to 1.0	2	8
Coarse Particles	1.0 to 20.0	1	4
		25 × 10^{-6} m^{-1}	100
San Manuel Smelter (62 km downwind) 10/4/77			
$(NH_4)_2SO_4$	0.1 to 1.0	41	43
SiO_2	0.1 to 1.0	9.5	10
Other Compounds	0.1 to 1.0	5.5	6
Coarse Particles	1.0 to 20.0	39	41
		95 × 10^{-6} m^{-1}	100
Mohave Power Plant (32 km downwind) 10/8/77			
$(NH_4)_2SO_4$	0.1 to 1.0	3	11
SiO_2	0.1 to 1.0	2	7
Other Compounds	0.1 to 1.0	1	4
Coarse Particles§	1.0 to 20.0	21	78
		27 × 10^{-6} m^{-1}	100

*Assumes that all fine particle sulfate exists as ammonium sulfate.
†Assumes that all fine particle silicon exists as SiO_2.
‡Given in TABLE 3.
§Determined from b_{scat} (total) − b_{scat} (fine particles).

The visibility budget for the Southwest background aerosol on October 9, 1977, determined from the Mie calculations, indicates that 52% of the light scattering was due to fine particles ($D_p \leq 1$ μm), 44% was due to Rayleigh scattering from gases, and 4% was due to coarse particles. Considering only the light scattering due to particles, 93% is due to fine particles. The detailed light scattering budget, summarized in TABLE 5, was constructed using the results of the fine particle mass balance and assumes that sulfur and silicon compounds have the same size distribution as the total fine particle aerosol. This is reasonable, since these compounds represent 82% of the measured fine particle mass.

The ratio of fine particle sulfate mass to the total b_{sp} value for the Southwest region, measured on October 9, 1977, is 0.15 g $(SO_4^=)$/m. A similar calculation for the fine particle SiO_2 mass/b_{sp} ratio yields 0.11 g (SiO_2)/m. Perhaps a more interesting calculation is the ratio of fine particle sulfate or SiO_2 mass to the contribution of that species to b_{sp}, as determined in the light scattering budget. This ratio is 0.4 g/m for both fine particle sulfate and SiO_2.‖ This calculation assumes that sulfur and silicon have the same size distribution.

‖The inverse of this ratio, i.e., the contribution of $SO_4^=$ or SiO_2 to b_{sp} divided by the fine particle $SO_4^=$ or SiO_2 mass, is (2.5 × 10^{-6} m^{-1})/(μg/m) for $SO_4^=$ and for SiO_2.

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Plume Impact

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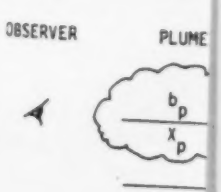

FIGURE 7. Schematic diag
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Plume Measurements

In order to characterize the emissions from specific sources in the region, measurements were made in plumes from the San Manuel copper smelter on October 1, 2, and 4, 1977, and the Mohave power plant October 8, 1977. Separate orbital flights in the plume and in the background, as well as plume traverses, were carried out at each distance downwind of the source.

To help evaluate the data presented in this section on source characterization, it is instructive to compare the emission rates of the San Manuel smelter and Mohave power plant. In 1975, the Mohave power plant emitted an average of 66 tons/day of SO_x and 77 tons/day of NO_x.[16] In 1977, the San Manuel smelter emitted 557 tons/day of sulfur expressed as SO_2.[17]

Plume Impact

The impact of a plume on visibility can be estimated by comparing the visual range with the plume present to the visual range through the background without the plume. The parameters used in this semiquantitative calculation are shown in FIGURE 7. The subscripts p and b indicate plume and background parameters, respectively. The light scattering coefficient in the plume, b, includes the background contribution.

The visual range, V, is defined as "the distance, under daylight conditions, at which the apparent contrast between a specified type of target and its background becomes just equal to the threshold contrast of an observer."[18] For the following analysis, the target and background are a black object viewed against the horizon sky, and the threshold contrast of a typical observer, 0.02, is used.[19] In this case, the visual range is given by:

$$V = \frac{-\ln 0.02}{\bar{b}_{ext}} = \frac{3.9}{\bar{b}_{ext}}$$

where $\bar{b}_{ext}$ is the average extinction coefficient along the sight path. This equation assumes uniform illumination along the sight path and neglects multiple scattering.

The following calculations assume that the contribution of absorption to extinction is small, so that the equation for visual range can be rewritten as $Vb_{scat} = 3.9$. For the case depicted above of a plume imbedded in background air, the corresponding

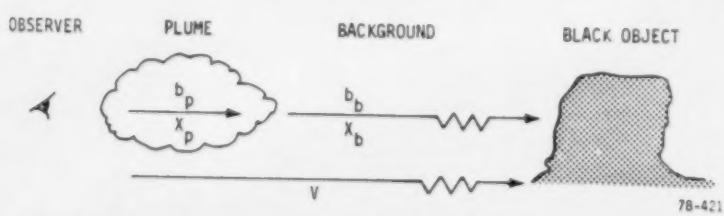

FIGURE 7. Schematic diagram of parameters used in semiquantitative calculation of plume impact. The subscripts p and b indicate plume and background parameters, respectively.

equation is:

$$X_p b_p + X_b b_b = 3.9$$

where b_p and b_b are the average scattering coefficients in the plume and background, respectively, and X_p is the measured plume width. The quantity $X_p b_p$, termed the optical depth of the plume (τ_p), is the integral of scattering across the plume, i.e., $\tau_p = \int b_p(x)dx$. The visual range looking through the plume was calculated using the equation:

$$V = X_p + X_b = X_p + \frac{3.9 - \tau_p}{b_b}$$

The values for X_p, b_p, and b_b were determined from measurements at the lowest altitude at which plume traverses were made. The plume opacity is defined in terms of the plume optical depth as $1 - \exp(-\tau)$.

The visual range and plume optics calculations are summarized in TABLE 6 and FIGURE 8. The visual range through the copper smelter plume is reduced at 8 km downwind of the stack from 135 km in the background to 13 km with the plume present (90% reduction). The visibility reduction decreases further downwind of the plant. At 60 km downwind, the visual range is 82 km with the plume present (39% visibility reduction relative to the background). The visibility reduction with the plume present is nearly equal at 60 and 127 km downwind of the smelter. The power plant plume exhibits quite different visual behavior. The reduction of the background visual range is less at 32 km than at 60 km downwind (11% vs. 25%, respectively). However, the power plant plume was measured under meteorological conditions that varied during the day, resulting in erratic plume behavior.

The difference in the reduction of the background visual range between these plumes may be the result of greater emissions released from the smelter stack and of greater dispersion of the Mohave plume on October 8.

TABLE 6

PLUME IMPACT DATA

	Downwind Distance (km)	Plume Width (km)	Plume Optical Depth	Plume Opacity	Visual Range		Visual Range Reduction due to Plume (%)
					With Plume (km)	Background	
San Manuel Smelter	8	16	4.9	0.99	12	135	90
	32*	20	2.8	0.94	54	135	60
	60	25	2.2	0.89	82	135	39
	127	54	3.1	0.95	78	135	42
Mohave Power Plant	32	8	0.66	0.48	98	110	11
	60	27	1.92	0.85	82	110	25

*Plume width at 32 km downwind, interpolated from measurements at 8 and 60 km downwind.

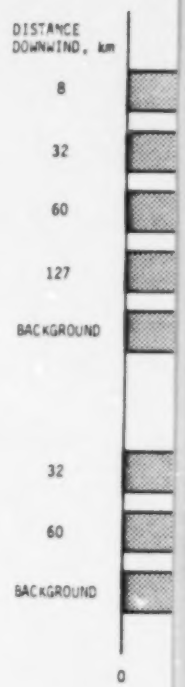

FIGURE 8. Visual ra
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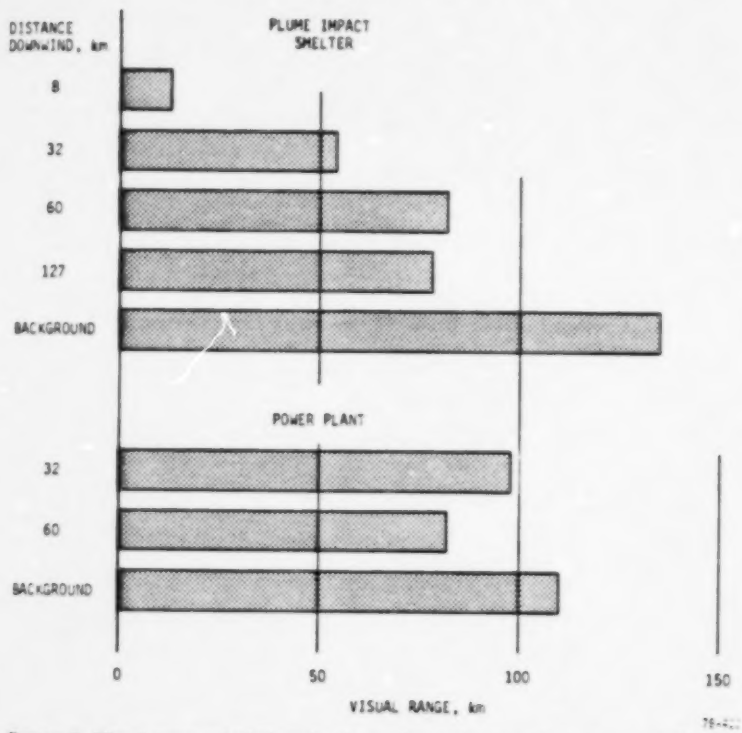

FIGURE 8. Visual range calculations for the smelter and power plant, at several distances downwind, compared to the background visual range.

Elemental Composition in Plumes

The elemental composition of the plume excess aerosol, i.e., the plume aerosol with the background subtracted, was determined from the analysis of samples collected in orbital plume flights. The background concentrations were determined in orbital flights upwind of the plume. The aerosol elemental compositions for the San Manuel smelter plume at 60 km downwind and the Mohave power plant at 32 km downwind are given in TABLE 7. The largest plume excess elemental constituents of the fine particle aerosol in both plumes are sulfur and silicon. Silicon, aluminum, calcium, and iron are major constituents of the plume excess coarse particles. These data indicate that coal-fired power plants and copper smelters in the Southwest may be major sources of the fine particle silicon as well as sulfur, the two major constitutents of the Southwest visibility-reducing aerosol.

Plume excess aerosol enrichment factors for the San Manuel smelter and Mohave power plant are given in TABLE 4. Coarse particle Si, K, Ca, Ti, and Fe in both plumes

TABLE 7
PLUME EXCESS* AEROSOL ELEMENTAL CONCENTRATION

Element†	San Manuel Smelter 60 km downwind (10/4/77)		Mohave Power Plant 32 km downwind (10/8/77)	
	Fine Particles‡ ($\mu g/m^3$)	Coarse Particles§ ($\mu g/m^3$)	Fine Particles ($\mu g/m^3$)	Coarse Particles ($\mu g/m^3$)
Al	<0.01	1.91	0.05	1.46
Si	1.08	6.12	0.34	3.67
S	2.29	—	0.30	—
K	0.43	0.60	—	0.25
Ca	—	1.58	—	1.16
Ti	0.02	0.03	—	0.04
Mn	—	0.02	—	—
Cr	0.20	0.06	—	0.05
Fe	0.02	1.02	—	0.55
Cu	—	0.06	—	—
Ni	—	—	0.20	0.01
Se	0.26	—	—	—
Pb	—	—	—	0.03

*Plume excess values were determined by subtracting background concentrations from plume concentrations of each element.

†Molybdenum was detected in concentrations above background but at the minimum detectable limit in the Mohave power plant plume.

‡Fine particle ($D_p \leq 1$ μm) concentrations were determined from the sum of impactor stage 4 and the final filter. Sulfur values were determined from TWOMASS samples.

§Coarse particle ($D_p \geq 1$ μm) concentrations were determined from the sum of impactor stages 1, 2, and 3.

and coarse particle Mn in the smelter plume are present in crustal abundances relative to Al. Thus, the crustal abundances of these elements in Southwest background coarse particles may be an indication that the source of these particles is crustal weathering or fly ash. Plume excess coarse particle Cr, Ni, Cu, and Pb are all enriched relative to crustal abundances. All plume excess fine particle species are enriched relative to the crust, except for fine particle Si in the Mohave plume.

The size distribution of plume excess elemental constituents shows many similarities with the regional data. Aluminum, calcium, and iron all exhibit the highest concentrations for particles with diameters >4 μm. Silicon has a size distribution similar to aluminum down to 1 μm but increases below that diameter. Sulfur is mainly concentrated in fine particles.

The total aerosol mass was not measured but can be estimated from the elemental concentrations and mean aerosol volume, as described previously. A comparison of these two approaches shows that in the San Manuel smelter on October 4, 1977, at 62 km downwind, 100% of the plume excess fine particle mass, determined from plume excess aerosol volume (assuming a mass-weighted density of 2 g/cm^3), can be accounted for from the measured constituents. The coarse particle volume was not measured in this plume. Approximately 73% of the plume excess fine particle mass is sulfate (expressed as ammonium sulfate), and 17% is silicon (expressed as SiO_2). In the Mohave power plant plume on October 8, 1977, only about 50% of the fine particle

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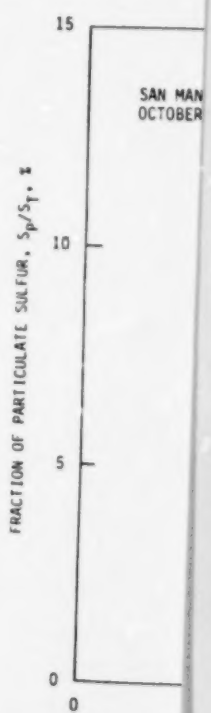

FIGURE 9. Determin
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to 1230 MST, between

mass determined from aerosol volume can be accounted for by the measured constituents. Of this 50% at 32 km downwind, plume excess sulfate [expressed as $(NH_4)_2SO_4$] accounts for 52% of the mass inferred from measured aerosol composition, and plume excess fine particle silicon (expressed as SiO_2) accounts for 31%.

Sulfur Transformation

The conversion of SO_2 to particulate sulfate in plumes can be estimated from the measurements of gaseous SO_2, particulate sulfate, light scattering coefficient, and mean accumulation mode volume. A number of methods to determine SO_2 conversion rates in power plant plumes have been used in the past.[20] The approach used here compares the fraction of particulate sulfur of the total sulfur, S_p/S_T, to plume age, estimated from analysis of winds aloft. Plume excess sulfur values with background

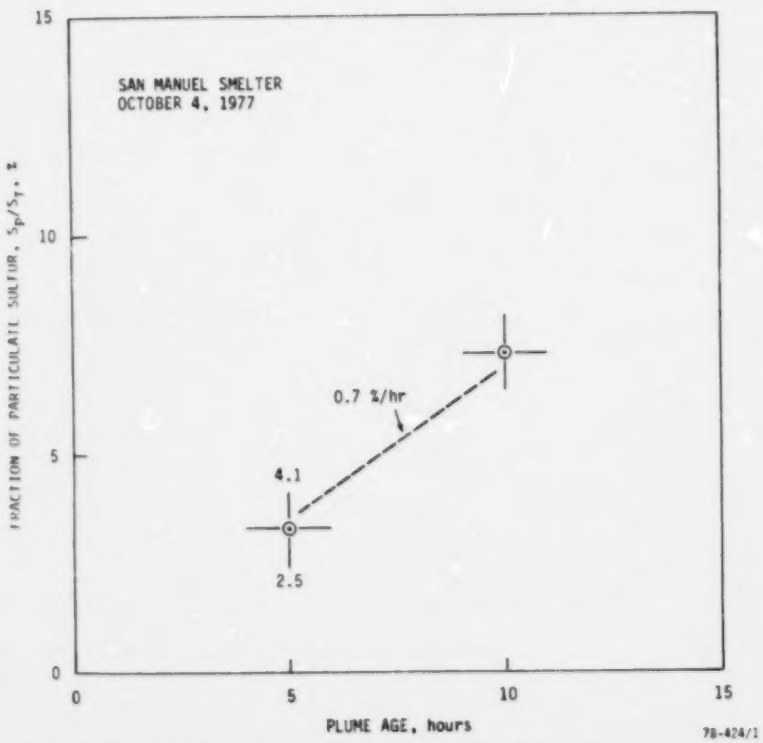


FIGURE 9. Determination of SO_2 conversion rate for the San Manuel smelter on October 4, 1977, from the fraction of particulate sulfur vs. plume age. Measurements were made from 0900 to 1230 MST, between 60 and 127 km from the plant.

subtracted were used in this calculation. The SO_2 conversion rate, measured in the San Manuel smelter plume on October 4, 1977, from 0900 to 1230 MST, was measured to be 0.7 ± 0.2%/hour between 60 and 127 km downwind from the plant (FIGURE 9). This calculation assumes no SO_2 deposition between measurements. However, even as much as 20% SO_2 deposition would not lower the conversion rate below the stated uncertainty. The SO_2 conversion rate was not calculated for the Mohave power plant plume because of poor plume resolution and erratic wind behavior encountered on October 8, 1977.

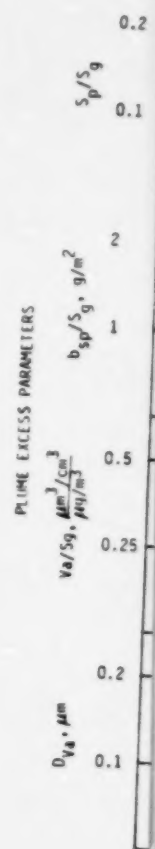
This single measurement of the $SO_2 \rightarrow SO_4^=$ conversion rate should not be considered typical without many additional measurements under a variety of conditions. It is interesting to note, however, that this value is within the range of conversion rates measured in plants in the Midwest.[21]

A number of other parameters can be used as qualitative indicators of SO_2 conversion, such as plume excess b_{sp}/S_g, S_p/S_g, and mean accumulation mode volume $(V_a)/S_g$ ratios, and accumulation mode aerosol size, D_{Va}. These indicators are shown graphically in FIGURE 10, as a function of distance downwind from the plant, for the San Manuel smelter and Mohave power plant plumes on October 4, 1977, and October 8, 1977, respectively. The three plume excess parameter ratios all increase downwind in both plumes, indicating that sulfur aerosol is being formed. The mean accumulation mode size decreases downwind from both plants, which also indicates fresh aerosol is being formed.

One method of assessing the effect of sulfur transformation on visibility is to determine the improvement of visual range if no SO_2 were converted to sulfate. This can be estimated for the San Manuel smelter at 20 and 32 km downwind on October 2, 1977, assuming that the b_{sp}/SO_2 ratio remains constant beyond 8 km downwind. The visual range with the plume present is then calculated, using b_{scat} and assuming no conversion, as described previously. This approach will slightly overestimate the effect of aerosol dilution (and therefore visual range) due to loss of SO_2 from the plume because of deposition and transformation. The results of this calculation are shown in FIGURE 11. At 32 km downwind from the smelter on October 2, the visual range would increase by nearly a factor of two if no SO_2 were converted to sulfate. At 60 km downwind, the visual range would be increased by 36% if there were no conversion. Thus, sulfur transformation has a large effect on visibility impairment within at least 60 km of the smelter.

Light Scattering Budget

The detailed light scattering contributions to visibility impairment as a function of particle size due to a smelter and power plant plume can be determined, using Mie calculations and measured plume excess aerosol size distributions, in an analogous manner to the treatment of regional data. However, there are several problems with the plume data. During the San Manuel smelter plume flights, only the fine particle size distribution was measured ($D_p \leq 1$ μm). During the Mohave power plant flights on October 8, 1977, the entire size distribution was measured, but windblown dust was occasionally mixing into the plume. Therefore, the plume visibility budgets must be considered as much more tentative than the regional visibility budget.



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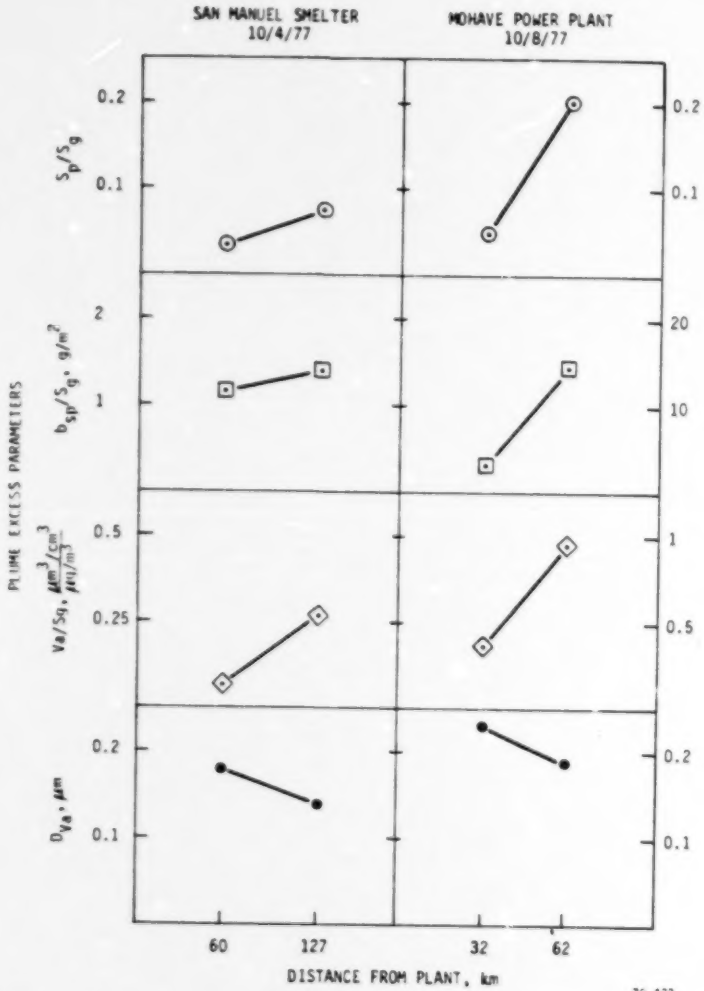

FIGURE 10. Qualitative indicators of SO_2 conversion in plumes, plotted vs. distance from plant.

In the San Manuel smelter plume at 62 km downwind on October 4, 1977, the plume excess b_{sp} value synthesized from only the measured fine particle size distribution ($D_p \leq 1$ μm), using Mie scattering functions, was $\Sigma b_{sp} = 56 \times 10^{-6}$ m^{-1}. This is 59% of measured value: b_{sp}(measured) = 95×10^{-6} m^{-1}. The detailed fine particle visibility budget given in TABLE 5 was determined from chemical composition

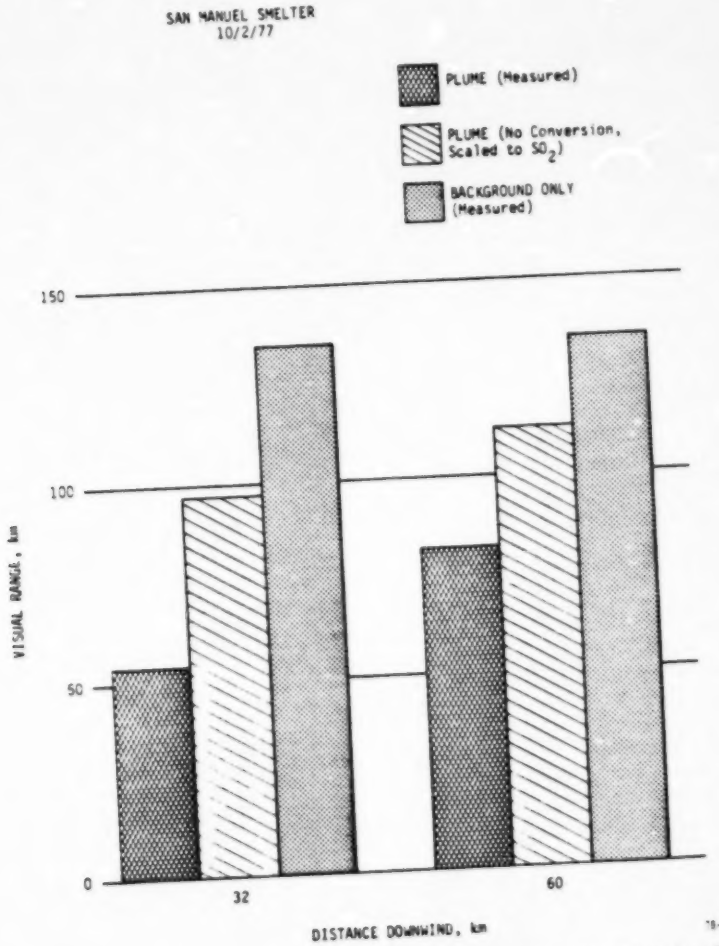

FIGURE 11. Visual range through the San Manuel smelter plume, with and without SO_2 conversion and through background only, for two downwind distances.

and size distribution, using Mie scattering calculations. Sulfate [assumed to be in the chemical form of $(NH_4)_2SO_4$] accounted for 43% of the total b_{sp} in the smelter plume of October 4, 1977; SiO_2 accounted for 10%.

The Mohave power plant plume excess b_{sp} calculation, on October 8, 1977, at 32 km downwind, was $\Sigma b_{sp}(\text{calc}) = 27 \times 10^{-6}$ m^{-1}, while the measured value was 18×10^{-6} m^{-1}. This difference may be due to the loss of large particles in the nephelometer sampling line.

Calculations of the light scattering budget for the Mohave power plant are complicated by the fact that aerosol composition calculations account for only about 50% of the fine particle aerosol mass as estimated from aerosol volume measurements. Therefore, the estimated contributions of individual chemical species to b_{sp} may be somewhat high. The visibility budget for the Mohave power plant plume, at 32 km downwind on October 8, 1977, is quite different from the smelter results. Coarse particles account for 78% of the calculated b_{sp}. This high value is at least partially due to windblown dust mixing into the power plant plume at the time of the measurement. Fine particle ammonium sulfate accounted for 11% of b_{sp} $\{b_{sp}[(NH_4)_2SO_4] = 3 \times 10^{-4}\ m^{-1}\}$. Fine particle silicon accounted for 7% of b_{sp} in the Mohave plant.

Summary

The atmospheric visibility-reducing aerosol in the Southwest has been experimentally characterized with respect to particle size, composition, and contribution to light scattering. Measurements were made on seven days within the mixing layer in the Southwest region, in the plume of a copper smelter (San Manuel smelter) and in the plume of a coal-fired power plant (Mohave power plant), using the MRI instrumented Beechcraft Queen Air aircraft. The aircraft was equipped to measure, and record on magnetic tape, the light scattering coefficient, Aitken nuclei count, size distribution, ozone, sulfur dioxide, nitrogen oxides, temperature, dew point, turbulence, pressure (altitude), and navigational parameters. Multistage impactor and size-fractionated filter samples were also collected, in order to determine aerosol elemental composition as a function of size. Visual range estimates were obtained by viewing distant landmarks and verified by optical photography.

Results of the study are summarized below:

1. On two regional flights over large parts of the Southwest, the visibility-reducing aerosol was quite homogeneous throughout the entire region, indicating that the visibility impairment was of regional extent.
2. The aerosol size distribution throughout the region on October 5 and 9, 1977, was bimodal, with a geometric volume mean size for the fine particles of ~0.25 microns, which is slightly smaller than the continental background aerosol average mean size (0.3 microns). The measured coarse particle mode volumetric mean size was ~5.5 microns, which is equal, within error, to the continental background.
3. The elemental size distribution, from both regional and plume impactor samples, indicated that aluminum, calcium, and iron are present predominantly in coarse particles; sulfur and titanium are present predominantly in fine partcles; and silicon and potassium have substantial concentrations in both modes.
4. Sulfur and silicon were found in nearly equal concentrations in the Southwest background data and were the elements present in the highest concentrations in the fine particles ($D_p < 1$ μm). The detailed aerosol mass balance determined for the Southwest region showed that sulfur (expressed as ammonium sulfate) and silicon (expressed as SiO_2) accounted to ~53% and ~29% of the fine particle mass, respectively.
5. A total of ~93% of the measured coarse particle ($D_p > 1$ μm) mass in the Southwest region was composed of elements that were present in the same abundances

relative to aluminum as in the earth's crust. This indicates that the source of these particles is either windblown dust or material with an elemental composition nearly that of crustal material, such as fly ash.

6. The light scattering budget for Southwest background aerosol on October 9, 1977, indicated that ~52% of the light scattering was due to fine particles, ~44% was due to Rayleigh scattering from gases, and ~4% was due to coarse particles. Considering only the light scattering due to particles, ~93% is due to fine particles that are composed mainly of sulfates [~53% as $(NH_4)_2SO_4$] and silicon compounds (~29% as SiO_2).

7. Mie scattering calculations of the light scattering coefficient due to particles, using the measured averaged regional size distribution, were in good agreement with the values measured with a nephelometer. This is a quantitative indication of the internal consistency of the size distribution and b_{scat} measurements, which adds credibility to the calculated light scattering budget.

8. Plume excess fine particle aerosol, i.e., the point source emission plume aerosol with the background subtracted, was composed largely of sulfur and silicon compounds for both the smelter and power plant plumes. The major elemental species in the plume excess coarse particle aerosol were aluminum, silicon, potassium, calcium, and iron, in approximately crustal abundances in both plumes. These results indicate that fine particle Si may be a good tracer for primary combustion aerosol in smelter and power plant plumes.

9. A simple semiquantitative calculation of visual plume impact is described that compares the visual range with and without the plume present. The smelter plume caused a ~90% reduction of visual range relative to the background visual range (135 km) at 8 km downwind from the plant. Increased b_{scat} and sulfate levels relative to background concentrations and a ~42% reduction of visual range were observed as far as 127 km downwind of the smelter plume on October 4, 1977. Measurements at 60 km downwind of the Mohave power plant plume on October 8, 1977, showed a ~25% reduction of visual range.

10. Sulfate aerosol was formed in Southwest power plant and smelter plumes. The measured SO_2 conversion rate from the San Manuel copper smelter on one morning between 0900 and 1230 (MST) was 0.7 ± 0.2%/hour between 60 and 127 km downwind.

11. The plume excess visibility budget indicated that fine particle sulfur and silicon species contributed ~53% to the excess b_{scat} in the San Manuel smelter plume. In the Mohave power plant plume, coarse particles were the major contributors to the excess b_{scat}, which may have been at least partially due to windblown dust on the sampling day.

Acknowledgments

We appreciate the guidance provided by William Wilson, Director of the Office of Regional Studies, and the field support provided by Tom Ellestad of EPA. The analysis herein was greatly enhanced by consultation with, and guidance from, S. K. Friedlander and R. Flagan of Caltech. We also appreciate the field support provided by J. A. McDonald, the data processing support of J. Roebuck, and the analysis

support of J. Ogren and G. Markowski. Part of the data analysis has been supported by the U.S. EPA under Grant No. R 802160 to Caltech.

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